

A review of life cycle greenhouse gas (GHG) emissions of commonly used *ex-situ* soil treatment technologies

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ABSTRACT

GHG emissions are important footprints because of increasing concern over climate change. Remediation methods produce GHGs in varying quantities based on which activities are involved through its entire life cycle and which contaminants are present, often because some contaminants are more difficult to remove from soil than others. Accounting for emissions from all phases of the project requires a life cycle assessment (LCA) approach. LCA can help in choosing the best available technology to reduce the environmental burden of the remediation technology or to improve the sustainability of the technology by implementing systematic approaches to ensure that future developments are optimized for environmental performance throughout the life cycle. The primary objective of this paper is to review existing LCA studies that report GHG emissions ($\text{CO}_2\text{-eq}$) or Global warming potential (GWP) from six *ex-situ* soil remediation technologies (ESRTs), including excavation and disposal, *ex-situ* thermal desorption, *ex-situ* soil vapor extraction, *ex-situ* bioremediation, excavation and incineration, and soil washing, and present the variability in GHG/GWP results and how this data can help in selecting an *ex-situ* soil remediation technology with a lower global warming potential. A second objective of this study is to compare the GWP levels of *ex-situ* remediation to the GWP levels of typical *in situ* remediation methods. Our results showed a large variation in GHG emissions of treated soil from six ESRTs varying from 3.1×10^{-7} t to 8.2 t $\text{CO}_2\text{-eq/m}^3$. Incineration had the highest mean GHG emissions (0.7 t $\text{CO}_2\text{-eq/m}^3$) and thermal desorption the lowest (0.07 t $\text{CO}_2\text{-eq/m}^3$). It was also found that there was a large variation range of GHG emissions from the *ex-situ* excavation and disposal method soil treatment technologies, varying from 3.1×10^{-7} t to 8.2 t $\text{CO}_2\text{-eq/m}^3$ of treated soil. This knowledge provides opportunities to increase sustainability of soil remediation throughout the investigation, design, construction, operation, and monitoring phases of site remediation regardless of the selected cleanup remedy. This shows data on LCA GHGs are useful to assess the impact of different scenarios and management practices on ESRTs.

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1. Introduction

Most systems and processes that require energy, emit greenhouse gases (GHG) and contribute to climate change (Raadal et al., 2011). Remediation activities often consume various chemicals, materials, and energy which are required for extraction, processing and transport of raw materials for use at remediation sites (Kim et al., 2013). Remediation techniques are often categorized as *in-situ* or *ex-situ* according to the removal or otherwise of the contaminated medium. *In situ* remediation does not require removal of contaminated soils and ground water. Therefore, this approach could be limited by its capacity to remove contaminated chemicals but may have lower costs of materials handling and environmental impacts (Lemming et al., 2010a). The *ex situ* remediation however involves excavation and extraction of soils and ground water for removal of contaminants. In some cases, the contaminated media may be moved to a treatment location (off-site treatment). This approach has advantages to select remediation techniques and therefore, is often effective in removing soil contaminants. However, the process uses extensive energy in excavation of contaminated soil for treatment either onsite, requiring less energy for transport or offsite, which most likely will require more energy for transportation of the contaminated soil to the treatment location. Amongst the different alternatives for site cleanup or remediation, the technology selected for the remediation amongst other factors may consider the kind of contaminants present at the site, the cleanup level, the time required for the remediation, economic resources, and the environmental impacts of the remediation process itself (Cappuyns, 2013a).

Any technique that remediates or manages the risk of contamination at a site has an environmental benefit as it eliminates or reduces exposure to contaminants. However, the remediation process or technique will introduce new environmental impacts due to the use of energy and materials, which cause emissions throughout their life cycle. These environmental impacts are often referred to as secondary impacts as opposed to the primary environmental impacts, which are related to the site, particularly the *ex-situ* remediation techniques (ESRTs) (Lemming et al., 2010a,b,c).

As Canada achieves its greenhouse gas emissions reduction target, activities carried out in each sector is important to contributing in achieving this goal. The waste management and remediation services in Canada contributed at least 3% of the total GHG emissions in 2015 (ECCC, 2017). GHG emissions are important footprints because of increasing concern over climate change (Kim et al., 2013). Generally, carbon dioxide (CO₂), nitrous oxide (N₂O), and methane (CH₄) contribute to GHG emission, and carbon dioxide (CO₂) is the primary indicator of the greenhouse effect (Kim et al., 2014; Unfccc, 2011; IPCC, 2006). Various soil remediation activities result in GHG emissions, including operation of equipment and energy use; transportation of personnel, materials, and

equipment; and the production of consumable materials. *Ex-situ* soil remediation will involve excavating large volumes of contaminated soil and transporting it to a proper treatment and/or disposal site whilst *in-situ* treatment involves the treatment of the contaminated soil medium in the site in which it was found (Lemming, 2018). In relation to GHG emissions, each remediation method can produce GHGs in varying quantities due to different activities involved through its entire life cycle to remove different types of contaminants. Because some contaminants are more difficult to remove from soil than others, they may require more equipment, chemicals, and consume more energy.

Accounting for emissions from all phases of the project requires a life cycle assessment (LCA) approach. Since the last decade, LCA has been gaining wider acceptance as a tool for the quantification of environmental impacts using global warming potential (GWP) which is a relative measure based on quantifying GHG emissions and evaluation of improvement options throughout the life cycle of a process, product or activity (Klöpffer and Grahl, 2014). In most industries such as waste management and remediation that use various technologies, LCA can help in choosing the best available technology to reduce the environmental burden of the remediation technology or to improve the sustainability of the technology by implementing systematic approaches to ensure that future developments are optimized for environmental performance throughout the life cycle (Morais and Delerue-Matos, 2010). Nunes et al. (2016) (Nunes et al., 2016) utilized LCA to compare treatment pathway alternatives applied to soil remediation with electrokinetic methods. Lemming et al. (2010a,b,c) evaluated environmental impacts of remediation of a chloroethene-contaminated site using LCA. However, to the best of our knowledge, no critical review using LCA has been performed to quantifying GHG emission and environmental impacts from *ex-situ* remediation techniques.

The primary objective of this paper is to review existing LCA studies that report GHG emissions from *ex-situ* soil treatment technologies. The secondary objective is to analyze the variability in GHG/GWP results and compare the GWP levels of *ex-situ* remediation to the GWP levels of typical *in-situ* remediation technologies (ISRTs). In addition, this study is to help good practice in selecting an ESRT with a lower GWP. This review is however, limited to the level of details that is provided on the models and data used in the LCA study. Furthermore, to provide a more comprehensive perspective of the environmental and total sustainability of ESRTs, other factors such as human health impacts, water consumption, jobs created, etc. have not been assessed.

2. Methodology

2.1. Search strategy and study evaluation

The methodology consisted of searching scientific databases for

peer-reviewed literature related to the use of LCA to estimate GHG emissions from *ex-situ* soil remediation technologies. The search focused on ESRTs often used for contaminated soil remediation. This included six general approaches identified as, 1) excavation and disposal, which involves removal of contaminated material (soil, solid wastes) from the contaminated site and transported to a permitted off-site treatment or disposal facility (Pavel and Gavrilescu, 2008), 2) *ex-situ* thermal desorption, which is a physical separation process where the contaminated soil is heated to volatilize water and organic pollutants (Federal Remediation Technologies Roundtable (FRTR), 2002), 3) *ex-situ* soil vapor extraction (SVE), in which case the excavated soil is often placed over a network of aboveground piping to which a vacuum is applied

such as CO₂ and CH₄ from soil remediation technologies addressed in numerous studies. Carbon dioxide equivalent (CO₂-eq.) and GWP is a standard unit for measuring other gas emissions, compared to the emissions of 1 ton of CO₂. The larger the GWP, the more that a given gas warms the Earth compared to CO₂ over that time period (Clune et al., 2017). Amongst the studies reviewed, GHG emissions were generally estimated based on Eq. (1). In order to compare the GHG emissions of the studies in this review, they were normalised to the volume of soil treated (t CO₂-eq/m³ of soil treated) if this was not already done in the study (see Eq. (1)). A few studies however reported GHG data per mass of soil treated. In those few cases, the volume was calculated from the density of soil (approximation based on moisture content).

$$\text{Normalised GHG emission} = \frac{\text{Total CO}_2 \text{ emissions throughout life cycle (tCO}_{2\text{eq}})}{\text{Volume of soil treated (m}^3\text{)}} \quad (1)$$

to encourage volatilization of organics (Hyman and Dupont, 2001), 4) *ex-situ* bioremediation, which usually involves placing excavated soil in a treatment area and aerated to enhance the degradation of organic contaminants by microbial population (Federal Remediation Technologies Roundtable (FRTR), 2002), 5) incineration, which utilizes high temperatures to volatilize and combust halogenated and other refractory organics in the contaminated soil (USEPA, 2012), and 6) soil washing (*ex-situ* soil separation processes), which is a water-based process for scrubbing soils *ex-situ* to remove contaminants either by dissolving or suspending them in the wash solution or by concentrating them into a smaller volume of soil through particle size separation, gravity separation, and attrition scrubbing (Raghavan et al., 1991). In few cases, the review was supplemented with literature from web-accessible documents and other grey literature. A systematic, staged search strategy was employed using the following search terms: '*ex-situ* soil remediation', '*in-situ* soil remediation', 'dig and dump' OR 'excavation and disposal' AND 'life cycle assessment/LCA' OR 'greenhouse gas/GHG' OR 'global warming potential/GWP' OR 'nitrous oxide/N₂O' OR 'methane/CH₄' OR 'carbon dioxide/CO₂' OR 'carbon footprint'.

2.2. LCA case studies included

This study reviewed almost 457 references, which were further refined to almost 140 reviewed papers that were associated to LCA from specific soil remediation technologies. Sixty-four of them were related to *ex-situ* soil remediation technologies applicable to oil and gas remediation. Only publications written in English with their original full texts available were included. Upon a closer examination of the full texts, only thirty-one (with 63 case studies, see Table 1) gave any form of quantitative estimation of lifecycle GHG emissions or at least reported on global warming impact. These 63 case studies mainly formed the primary basis of this review, although other publications within the database were used. Table 2 presents the number of case studies reviewed per ESRT.

2.3. Greenhouse gas emissions included

The focus of this review is on GHG emissions, although climate change is only one of several important environmental impacts when assessing different soil remediation technologies. Again, despite the fact that there are other environmental emissions (e.g., NO_x and SO₂) (IPCC, 2006), this review focuses on GHG emissions,

3. Life cycle assessment of remediation technologies

The LCAs in this study have been reviewed using a variety of methods, assumptions, databases and software. Indeed, the application of LCA to site remediation is an opportunity for site managers to make objective comparisons between several other alternatives (Onwubuya et al., 2009). Some remediation techniques require a lot more energy but for a shorter period of time while other technologies are required for a longer period but consumes much less energy. LCA provides a tool to initiate such a comparison of the various techniques based on the needs of the stakeholders involved. However, the LCA methodology is not perfect and still faces some challenges. Some researchers (Lemming et al., 2010a), (Suér et al., 2004), (Owsianiak et al., 2013) have pointed out some challenges with the LCA methodology and its application to remedial activities and raised concerns about conclusions made from most of these studies. Most of these discrepancies arise due to the different specific considerations for each study since remedial actions heavily depend on specific LCA methodology and assumptions made on the inventory data. Although none of these investigators was able to offer solutions to critically improve the existing LCA methodology, they generally have consensus about the methodological issues of LCA, and agree that the use of an inadequate methodology may lead to misleading conclusions to the practitioners and decision-makers in the field of soil and ground-water remediation.

4. Global warming potential (kg CO₂-eq.) as decision parameter for selection of remediation technology

Increasing concerns regarding global warming (climate change) have prompted major efforts across the globe to reduce GHG emissions caused by various activities such as fossil fuel consumption (USEPA, 2008a). Accordingly, one life cycle impact category receiving considerable attention in practices for green and sustainable remediation, and gradually becoming important in comparing the sustainability of remediation technologies is global warming impact (potential). Identifying stages within the life cycle of a specific remediation technology contributing the most to global warming, provides the opportunity to develop strategies and approaches to reduce energy consumption and GHG emissions.

Table 1

Overview of the LCA studies considered in the present study.

Case study	Ex situ remediation/Treatment method or technology	Contaminant type	Country/region	Comments
Ambrosini et al. (2014)	Excavation and disposal	Hydrocarbons	Italy	Backfilling after biopile treatment
Ambrosini et al. (2014)	Bioremediation	BTEX	Italy	
Beames et al. (2015)	Thermal desorption	Heavy metals	Belgium	
Beinat et al. (1998)	Excavation and disposal	Chlorinated solvents	Netherlands	14.3 tonnes per capita, also expressed per diesel consumption (0.077133 kg/MJ)
Boesch et al. (2014)	Incineration	Slag, fly ash	Switzerland	
Boesch et al. (2014)	Excavation and disposal	Slag, fly ash	Switzerland	
Busset et al. (2012)	Incineration	PCBs	France	600 tonnes of soil to be treated
Busset et al. (2012)	Bioremediation	PCBs	France	Electrical aeration; 600 tonnes of soil to be treated
Busset et al. (2012)	Bioremediation	PCBs	France	Mechanical aeration; 600 tonnes of soil to be treated
Cadotte et al. (2007)	Bioremediation	Diesel oil	Canada	Use of biopile sheets
Cappuyns (2013a,b)	Excavation and disposal	Petroleum hydrocarbon (HC)	Netherlands	Industrial site
Cappuyns (2013a,b)	Excavation and disposal	Petroleum hydrocarbon (HC)	Netherlands	Abandoned oil and gas processing plant
Cappuyns (2013a,b)	Excavation and disposal	Petroleum hydrocarbon (HC)	Netherlands	Distribution center for new and second-hand cars
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 50 mg/kg
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 100 mg/kg
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 255 mg/kg
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 400 mg/kg
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 500 mg/kg
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 1500 mg/kg
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 3000 mg/kg
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 6000 mg/kg
Choi et al. (2016)	Excavation and disposal	PCBs, PAHs, chlorinated pesticides	USA	Criteria of 9000 mg/kg
Diamond et al. (1999)	Excavation and disposal	General	Canada	75 weeks, 25 yrs of longer term impacts; included decontamination and demolition of existing buildings onsite; 14973 m ³ of soil to be treated
Ferdos and Rosen (2013)	Excavation and disposal	Heavy metals	Sweden	Use of sitewise tool; 100 km to disposal site by truck
Ferdos and Rosen (2013)	Excavation and disposal	Heavy metals	Sweden	2 km to disposal site by truck
Ferdos and Rosen (2013)	Excavation and disposal	Heavy metals	Sweden	265 km to disposal site by train
Ferdos and Rosen (2013)	Excavation and disposal	Heavy metals	Sweden	270 km by boat + 21 km by truck
Ferdos and Rosen (2013)	Soil vapor extraction	Heavy metals	Sweden	20 yrs of monitoring & wastewater treatment
Ferdos and Rosen (2013)	Soil washing	Heavy metals	Sweden	270 km by boat + 21 km by truck
Ferdos and Rosen (2013)	Soil washing	Heavy metals	Sweden	100 km by truck to disposal site
Ferdos and Rosen (2013)	Soil washing	Heavy metals	Sweden	265 km by train to disposal site
Gill et al. (2016)	Soil vapor extraction	Hydrocarbons	United Kingdom	Equipment manufacture not accounted for; 9000 m ³ of soil treated
Gill et al. (2016)	Bioremediation	Hydrocarbons	United Kingdom	Equipment manufacture not accounted for; 9000 m ³ of soil treated
Godin et al. (2004a,b)	Excavation and disposal	Spent pot lining (SPL)	Canada	
Harbottle et al. (2006, 2007)	Excavation and disposal	BTEX and PTH	UK	
Hou et al. (2016)	Thermal desorption	Mercury	China	
Hou et al. (2017)	Excavation and disposal	Lead (Pb)	USA	
Hu et al. (2011)	Incineration	Polychlorinated Biphenyl (PCB)	USA	
Inoue and Katayama (2011)	Bioremediation	POPs	Japan	Landfarming; 220 m ³ per day for 180 days
Inoue and Katayama (2011)	Bioremediation	POPs	Japan	Use of biopile sheets; 220 m ³ per day for 180 days
Inoue and Katayama (2011)	Excavation and disposal	POPs	Japan	220 m ³ per day of soil treated for 15 days
Inoue and Katayama (2011)	Thermal desorption	POPs	Japan	3 metric tonnes soil per hour for 32 days
Kim et al. (2013)	Soil washing	Lead (Pb)	South Korea	4275m ³ of soil to be treated
Lemming et al. (2010a,b,c)	Excavation, treatment and disposal	Chlorinated ethenes	Denmark	1120 m ³ of soil treated off-site
Lim et al. (2016)	Bioremediation	TPH and BTEX	South Korea	Landfarming; 12 months duration; 55293 m ³ of soil treated
Loranger and Bage (2011)	Excavation and disposal	PAHs	Canada	
McAlexander and Tuggle (2015)	Excavation and disposal	Petroleum hydrocarbon (HC)	USA	
Page et al. (1999)	Excavation and disposal	Lead (Pb)	Canada	75 weeks; 1.2 × 10 ¹⁰ m ³ ; 0.3 m depth
Reddy and Chirakkara (2013)	Excavation and disposal	Slag, fly ash	USA	
Sanscartier et al. (2010)	Bioremediation	Petroleum hydrocarbon (HC)	Canada	Off-site biopile treatment

(continued on next page)

Table 1 (continued)

Case study	Ex situ remediation/Treatment method or technology	Contaminant type	Country/region	Comments
Sanscartier et al. (2010)	Bioremediation	Petroleum hydrocarbon (HC)	Canada	On-site biopile treatment
Suer and Andersson-Sköld (2011)	Excavation and disposal	Petroleum hydrocarbon (HC)	Sweden	
Toffoletto et al. (2005)	Bioremediation	Petroleum hydrocarbon (diesel)	Canada	Biopile treatment; Target = 700 mg HC/kg soil; 2 yrs treatment period
US Airforce and USEPA (2012) (Mascoet al., 2013)	Excavation and disposal	PAHs, TPH, PCBs, dioxins	USA	Includes off-site waste management
US Airforce and USEPA (2012) (Mascoet al., 2013)	Excavation and disposal	PAHs, TPH, PCBs, dioxins	USA	Does not include waste management off-site
Vignes (2001)	Thermal desorption	Various contaminants	USA	
Vocciant et al. (2016)	Bioremediation	Zinc	Italy	Area of 48 m ² ; 3.5 m depth
Volkwein et al. (1999)	Excavation and disposal	PAH, mineral oil, chromium	Germany	
Volkwein et al. (1999)	Soil washing	PAH, mineral oil, chromium	Germany	
Weiss Associates (2012)	Excavation and disposal	PCBs, PAHs	USA	On-site; include ground water monitoring
Weiss Associates (2012)	Excavation and disposal	PCBs, PAHs	USA	On-site; include existing infrastructure demolition
Weiss Associates (2012)	Excavation and disposal	PCBs, PAHs	USA	Off-site disposal; long term groundwater monitoring
Weiss Associates (2012)	Excavation and disposal	PCBs, PAHs	USA	

4.1. Determining the global warming impact (kg CO₂ eq.)

To determine the global warming impact, one has to first calculate the GHG emissions from each contributing source. The Intergovernmental Panel on Climate Change (IPCC) provided Guidelines for estimating GHG emissions (IPCC, 2006). The guidelines consist of five different volumes and include new sources and gases as well as updates to previously published methods (Environment and Climate Change Canada, 2016). Often, GHG emissions are calculated by multiplying the amount of fuel (diesel) or electricity consumed for example by the emission factor (EF) of each associated gas (CO₂, CH₄ and N₂O in this case). The default EF values as Tier 1 approach can be obtained from the IPCC reports (2006). Eq. (2) describes how each greenhouse gas can be determined from activities contributing to GHG emissions according to (Maciel et al., 2015).

$$GHG_i = \left(\sum C \times EF_i \right) \quad (2)$$

where GHG_i (greenhouse gas emissions); EF_i is the emission factor for the corresponding gas, i ; C is the amount of fuel or electricity consumed. Since the values of GHG emissions cannot simply be added due to their different effects on global warming, their values must be converted to common units by applying global warming potentials (GWPs). GWPs allow comparisons of the ability of each greenhouse gas to trap heat in the atmosphere relative to carbon dioxide (CO₂) over a specified period (Goglio et al., 2017). As such, the results of applying GWPs are expressed in carbon dioxide

equivalent (kg CO₂ eq.). Since each of the emissions is now in a common unit, they can be summed up to provide a single value representing the GHG emissions resulting from the specified activity (e.g. fuel or electricity consumption). After the summed GWPs are normalized using Eq. (1), we can compare the climate change impact of different remediation technologies and help them to make the most informed decision.

4.2. Current status of technology per GHG emissions

At the moment, new soil remediation technologies are been developed to have higher removal efficiencies and lower GHG emissions (Kuppusamy et al., 2016). However, the origin and nature of diesel extraction for example, that is required for excavation equipment and other transportation needs during the entire life cycle of the remediation project could strongly influence the overall global warming impact of the new technology. The change in upstream extraction approaches may have contributed as much to the environmental benefit as the change in actual remediation technology. As such, technology improvement must be a combination of actual technological updates as well as devising cleaner ways for upstream extraction of materials and energy. Therefore, most of these new technologies are developing technologies that are not in commercial operation as yet (Hou et al., 2014). Mature systems are those that are currently commercially available, including the six technologies reviewed in this study. Here, the data estimates of GHG emissions are much more reliable and commercially available ESRTs as compared to systems under development.

Table 2

List of studies reviewed per technology.

Ex-situ soil remediation technology	Number of case studies	References
Soil washing	6	(Kim et al., 2013), (Volkwein et al., 1999), (Ferdos and Rosen, 2013)
Incineration	3	(Boesch et al., 2014), (Busset et al., 2012), (Hu et al., 2011)
Bioremediation	12	(Ambrosini et al., 2014), (Sanscartier et al., 2010), (Cadotte et al., 2007), (Busset et al., 2012), (Gill et al., 2016), (Inoue and Katayama, 2011), (Lim et al., 2016), (Vocciant et al., 2016)
Excavation and disposal	36	(Ambrosini et al., 2014), (Diamond et al., 1999)– (Volkwein et al., 1999), (Associates, 2012), (Ferdos and Rosen, 2013), (Beinat et al., 2015), (Boesch et al., 2014), (Cappuyns, 2013b), (Godin et al., 2004b), (Hou et al., 2017), (Inoue and Katayama, 2011), (Lemming et al., 2010b), (McAlexander and Tuggle, 2015; Page et al., 1998; Reddy and Chirakkara, 2013; Suer and Andersson-Sköld, 2011), (Harbottle et al., 2006) (Loranger and Bage, 2011) (Mascoet al, 2013)
Thermal desorption	4	(Beames et al, 2015), (Hou et al., 2016), (Inoue and Katayama, 2011), (Vignes, 2001)
Soil vapor extraction	2	(Ferdos and Rosen, 2013), (Gill et al., 2016)
Total	63	

5. Results and discussion

This section presents the GHG emissions from the six ESRTs including excavation and disposal, *ex-situ* thermal desorption, *ex-situ* soil vapor extraction (SVE), *ex-situ* bioremediation, excavation and incineration, and soil washing (*ex-situ* soil separation processes). Results are based on 63 LCAs, published between 1997 and 2017 (Table 1). Every attempt has been made to identify criteria, considerations and assumptions leading to the greenhouse gas estimates for the respective ESRT.

5.1. GHG emissions of case studies

In order to implement a fair comparison, all GHG emissions data were normalized using Eq. (1) per volume of contaminated soil treated (Table 3).

Table 3 also lists the type of remediation technology, the contaminant type and the volume of soil treated. Fig. 2 shows the mean, minimum and maximum values of GHG emission for each type of remediation technology (SVE, thermal desorption, excavation and disposal, bioremediation, incineration and soil washing). The applied approach for estimation of GHG emissions is rather similar in the studies.

5.2. Variability of GHG emissions from LCA studies

There are different reasons for variability in LCA studies. It is found that the differences exist in soil and groundwater remediation technology GHG estimates. This can be attributed to a variety of causes, such as geographical and technological scope of modelling, functional unit and comparability, assumptions made on upstream impact, energy consumption, LCA software used, etc. Some of these causes are discussed below.

5.2.1. Technological and geographical scope

As a review with meta-analysis, it is difficult to investigate the source of the GHG emissions and a full understanding of the practical site systems. This may affect the GHG values generated in the LCAs. To enable a fair, accurate and correct comparison between studies, we carefully identified and documented the reference year and the geographical origin of the materials, equipment and chemicals used for the soil treatment, since these pieces of data could significantly influence the outcome. As reported in Fig 2, the range of emission data represents several technological and geographical scopes of the various LCA studies. The estimated GHG values were mainly based on energy consumptions from the six ESRTs. It can be seen that GWPs from incineration, extraction and disposal and vapor extraction are top three in the six ESRTs. The highest mean GHG emission value, $0.7 \text{ t CO}_2\text{-eq/m}^3$ is associated with incineration, whilst the lowest, $0.07 \text{ t CO}_2\text{-eq/m}^3$ is associated with thermal desorption. This can be explained because the incineration had the dominated energy consumption due to high temperature heating and incineration of the contaminated soils. The excavation and disposal approach required usually big equipment, such as digger and track transportation, leading to a higher fuel consumption. Soil vapour extraction used contaminant phase changes through mass transfer to collect the gas phase contamination at extraction wells. Although its operating temperature is lower than incineration, it is still an approach using high temperature heating up to contaminant gasification, leading to a higher energy consumption. It can also be found that there is a large variation in GHG emissions from the *ex-situ* excavation and disposal method soil treatment technologies, varying from $3.1 \times 10^{-7} \text{ t}$ to $8.2 \text{ t CO}_2\text{-eq/m}^3$ of treated soil. This is because the contaminated soil treatment using the excavation and disposal

could vary greatly. If using incineration and a long distance transport, its GWP could rise to a high level. However, if using the on-site thermal desorption or soil washing, the energy consumption could be reduced significantly, leading to a lower GWP.

For offsite treatments, the distance of soil treated from the site in the life cycle of the treatment is a key factor in the choice between in-situ and ex-situ offsite treatment since a long distance from its site requires more energy for the transport. Ambrosini et al., 2014 calculated GHG emissions for the remediation of gas stations at different geographical locations for Europe. Although the same treatment technology (excavation and disposal) was used in all cases, due to differences in transportation distances of materials, equipment and chemicals used, the calculated GHG emissions from the treatment process were different for each remediated site. Since remediation technologies are developing rapidly with some attention to low carbon emission technologies, GHG emissions data sets in already established life cycle inventory (LCI) databases may require regular updates. However, significant discrepancies exist among various LCI datasets, even if for only electricity technologies. Many factors can affect LCI datasets, such as remediation methods, local conditions, system boundaries and data sources. Therefore, some LCI datasets might be incoherent (Ambrosini et al., 2014). As such, relying on outdated life cycle inventory data can be misleading. Recent and up-to-date data should be used to obtain consistent and realistic LCI data (Turconi et al., 2013).

5.2.2. Functional unit and comparability

The selected functional unit of an assessment provides the possibility for comparability across studies. This makes the functional unit an extremely critical step in the LCA study. Some LCA studies such as Diamond et al., 1999, recommend that “the functional unit should relate to the production of an amount of treated soil”. However, a significant number of site remediation LCA studies (as seen in Table 1) also consider the treatment of an amount of soil or groundwater as the functional unit. For a study to be comparable within the same functional unit, independent technologies should provide the exact same service to the system (Turconi et al., 2013). There are instances where one study considers both primary and secondary impacts. Primary impacts refer to the local impacts associated with residual contamination left after remediation, whilst secondary impacts are related to the resource use and emissions arising from the treatment process (Lemming et al., 2010a). In a case where both impacts are included, the determined GHG emissions include emissions from both primary and secondary impacts. However, if either secondary or primary impact is included, this will be representative of the GHG emissions determined. This makes GHG emissions comparability challenging especially if details are not provided on what is included in the total GHG emissions calculated. For example, Choi et al, 2016 evaluates secondary environmental impacts of various remedial alternatives whilst Volkwein et al., 1999, considers both primary and environmental impacts in the study. It is important therefore, that one understands fully the context of such emissions determination in order to use GHG emissions result in any future work.

5.2.3. Contributions from upstream impacts

Results from this literature review indicate that it is important to correctly set the system boundaries to avoid the undesired cut-off of a relevant part of the emissions. Page et al., 1999 included the extraction of diesel used by equipment and machinery on-site in the system boundary, whilst Volkwein et al., 1999 does not include this. This important difference reflects significantly in the magnitudes of CO_2 equivalent emissions. Again, although transportation emissions usually represent a significant percentage of

Table 3
Overview of GHG (t CO_{2eq}) of ESRTs.

Case study	Technology	Volume of treated soil (m ³)	Contaminant type	GHG/GWP (kg CO ₂ -eq/site)	Normalized GHG/GWP (t CO ₂ -eq. Per m ³ of soil treated)
Ambrosini et al. (2014)	Excavation and disposal	6300	Hydrocarbons	2.67E+03	4.24E-04
Ambrosini et al. (2014)	Bioremediation	420	BTEX	9.74E+02	2.32E-03
Beames et al. (2015)	Thermal desorption	74000	General	1.57E+07	2.12E-01
Beinat et al. (1998)	Excavation and disposal	14.3	Chlorinated solvents	1.43E+01	
Boesch et al. (2014)	Incineration		Slag, fly ash	2.32E+03	
Boesch et al. (2014)	Excavation and disposal		Slag, fly ash	2.25E+03	
Busset et al. (2012)	Incineration	600	PCBs	6.50E+05	2.08E+00
Busset et al. (2012)	Bioremediation	600	PCBs	7.20E+04	2.30E-01
Busset et al. (2012)	Bioremediation	600	PCBs	7.50E+04	2.40E-01
Cadotte et al. (2007)	Bioremediation	2880	Diesel oil	3.60E+06	1.25E+00
Cappuyns (2013a,b)	Excavation and disposal	14000	Petroleum hydrocarbon (HC)	1.20E+06	8.57E-02
Cappuyns (2013a,b)	Excavation and disposal	80000	Petroleum hydrocarbon (HC)	5.20E+06	6.50E-02
Cappuyns (2013a,b)	Excavation and disposal	6150	Petroleum hydrocarbon (HC)	2.20E+04	3.58E-03
Choi et al. (2016)	Excavation and disposal	6241	PCBs, PAHs, chlorinated pesticides	7.47E+05	1.20E-01
Choi et al. (2016)	Excavation and disposal	4786	PCBs, PAHs, chlorinated pesticides	6.09E+05	1.27E-01
Choi et al. (2016)	Excavation and disposal	2896	PCBs, PAHs, chlorinated pesticides	4.30E+05	1.48E-01
Choi et al. (2016)	Excavation and disposal	1673	PCBs, PAHs, chlorinated pesticides	3.15E+05	1.88E-01
Choi et al. (2016)	Excavation and disposal	766	PCBs, PAHs, chlorinated pesticides	2.28E+05	2.98E-01
Choi et al. (2016)	Excavation and disposal	375	PCBs, PAHs, chlorinated pesticides	1.91E+05	5.09E-01
Choi et al. (2016)	Excavation and disposal	212	PCBs, PAHs, chlorinated pesticides	1.75E+05	8.25E-01
Choi et al. (2016)	Excavation and disposal	165	PCBs, PAHs, chlorinated pesticides	1.36E+05	8.24E-01
Choi et al. (2016)	Excavation and disposal	99	PCBs, PAHs, chlorinated pesticides	8.16E+05	8.24E+00
Diamond et al. (1999)	Excavation and disposal	14973	General	3.77E+06	2.52E-01
Ferdos and Rosen (2013)	Excavation and disposal	280000	Heavy metals	6.20E+06	2.21E-02
Ferdos and Rosen (2013)	Excavation and disposal	280000	Heavy metals	4.70E+06	1.68E-02
Ferdos and Rosen (2013)	Excavation and disposal	280000	Heavy metals	3.60E+06	1.29E-02
Ferdos and Rosen (2013)	Excavation and disposal	280000	Heavy metals	1.20E+07	4.29E-02
Ferdos and Rosen (2013)	Soil vapor extraction	280000	Heavy metals	4.20E+06	1.50E-02
Ferdos and Rosen (2013)	Soil washing	280000	Heavy metals	3.80E+06	1.36E-02
Ferdos and Rosen (2013)	Soil washing	280000	Heavy metals	3.60E+06	1.29E-02
Ferdos and Rosen (2013)	Soil washing	280000	Heavy metals	3.90E+06	1.39E-02
Gill et al. (2016)	Soil vapor extraction	900	Hydrocarbons	3.80E+05	4.22E-01
Gill et al. (2016)	Bioremediation	900	Hydrocarbons	5.00E+05	5.56E-01
Godin et al. (2004a,b)	Excavation and disposal	542500	Spent pot lining (SPL)	9.12E+03	1.68E-05
Harbottle et al. (2006)	Excavation and disposal	4400	BTEX and PTH	1.29E+01	2.93E-03
Hou et al. (2016)	Thermal desorption	10000	Mercury	3.57E+02	3.57E-02
Hou et al. (2017)	Excavation and disposal	1270	Lead (Pb)	8.16E+05	6.43E-01
Hu et al. (2011)	Incineration	34000	Polychlorinated Biphenyl (PCB)	4.32E+02	2.44E-02
Inoue and Katayama (2011)	Bioremediation	220	POPs	1.57E+00	7.14E-03
Inoue and Katayama (2011)	Bioremediation	220	POPs	1.80E+00	8.18E-03
Inoue and Katayama (2011)	Excavation and disposal	220	POPs	8.70E+00	3.95E-02
Inoue and Katayama (2011)	Thermal desorption	3	POPs	7.13E+00	1.78E-02
Kim et al. (2013)	Soil washing	4275	Lead (Pb)	5.01E+05	1.17E-01
Lemming et al. (2010a,b,c)	Excavation and disposal	1120	Chlorinated ethenes	1.20E+05	1.07E-01
Lim et al. (2016)	Bioremediation	55293	TPH and BTEX	5.16E+02	9.33E-06
Loranger and Bage, 2011	Excavation and disposal	15000	PAHs	82.5	5.50E-03
McAlexander and Tuggle (2015)	Excavation and disposal	720	Petroleum hydrocarbon (HC)	70	9.72E-02
Page et al. (1999)	Excavation and disposal	1.20E+10	Lead (Pb)	3.70E+06	3.08E-07
Reddy and Chirakkara (2013)	Excavation and disposal	57726.9	Slag, fly ash	1.00E+04	1.73E-04
Sanscartier et al. (2010)	Bioremediation	112	Petroleum hydrocarbon (HC)	1.49E+04	1.33E-01
Sanscartier et al. (2010)	Bioremediation	112	Petroleum hydrocarbon (HC)	7.26E+03	6.48E-02
Suer and Andersson-Sköld (2011)	Excavation and disposal	6500	Petroleum hydrocarbon (HC)	2.80E+05	4.31E-02
Toffoletto et al. (2005)	Bioremediation	8000	Petroleum hydrocarbon (diesel)	6.20E+05	7.75E-02
US Airforce and USEPA (2012)	Excavation and disposal	31200	PAHs, TPH, PCBs, dioxins	660	2.12E-02
US Airforce and USEPA (2012)	Excavation and disposal	31200	PAHs, TPH, PCBs, dioxins	170	5.45E-03
Vignes (2001)	Thermal desorption	25254	Various contaminants	2.38E+04	9.42E-04
Vocciant et al. (2016)	Bioremediation	28224	Zinc	5.63E+02	1.99E-05
Volkwein et al. (1999)	Excavation and disposal	1810	PAH, mineral oil, chromium	6.95E+04	3.84E-02
Volkwein et al. (1999)	Soil washing	1505	PAH, mineral oil, chromium	3.77E+05	2.50E-01
Weiss Associates (2012)	Excavation and disposal	40300	PCBs, PAHs	3.95E+06	9.80E-02
Weiss Associates (2012)	Excavation and disposal	274700	PCBs, PAHs	5.24E+06	1.91E-02
Weiss Associates (2012)	Excavation and disposal	117200	PCBs, PAHs	1.44E+07	1.23E-01
Weiss Associates (2012)	Excavation and disposal	123400	PCBs, PAHs	1.52E+07	1.23E-01

the total GHG emissions from off-site *ex-situ* soil remedial options, some studies have shown that they may be less relevant for other systems (i.e., most *in-situ* and *ex situ* on-site remedial applications). Fig. 4 shows contributions from the main input activities from different cases of soil remediation involving excavation based on

the studies reviewed that had reported contributions per major process inputs. In these studies, as shown in Fig. 3, transportation is quite significant in the entire life cycle of the remediation processes. Upstream processes potentially having relevant impacts include fuel provision and the commissioning and

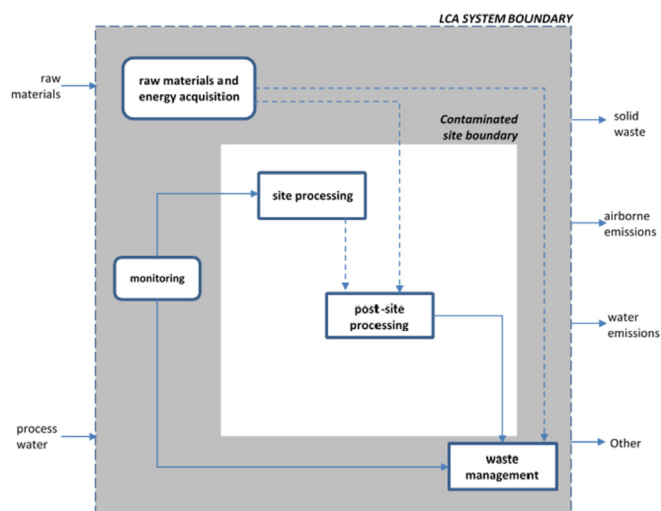


Fig. 1. Typical site remediation LCA.

decommissioning of site machinery. According to Amponsah et al., 2014), few authors consider fuel provision negligible with regards to GHG emissions (e.g., (Meyer-Aurich et al., 2012)), while others depict it as important (e.g., (USEPA, 2012), (Owsianiak et al., 2013), (Rivett et al., 2002; Godin et al., 2004a; USEPA, 2008b; Wang, 2007)). Results from these studies show that, assumptions made about contributions from upstream processes such as material and chemical extraction can affect conclusions drawn from a study. As an example, infrastructure (construction works, aeration systems, nutrient systems and leachate collection systems) usually required based on the technology for on-site or off-site treatment was considered a minor issue in some studies (Associates, 2012)—(Ferdos and Rosen, 2013), while it was found to be more relevant (in some cases dominant) (Ambrosini et al., 2014), (Associates, 2012), (Sanscartier et al., 2010), (Cadotte et al., 2007).

5.2.4. LCA software and inventory

The particular LCA software and inventory utilized in a particular study can be a major cause for variation in the CO₂-equivalents reported. In fact, recent LCA studies rely on a variety of different commercial software including different versions of SimaPro

(simapro manual PRE Consultants, 2008) and GaBi (Herrmann and Moltesen, 2014), as well as different life cycle inventory data, such as EcoInvent Database or in some cases, site-specific data. Some studies relied on public domain footprint analysis tools such as SiteWise™ (US Naval Facilities Engineering Command, 2011), Sustainable Remediation Tool (SRT™) (AFCEE, 2010), and the U.S. EPA's Spreadsheets for Environmental Footprint Analysis (SEFA) (Huang et al., 2016). Again, other studies do not say explicitly which data or software is used in the analysis e.g. (Scheuermann, 2009). Indeed, the differences in data sets associated with LCA inventories for even similar processes or products impact the GHG emissions estimated. Large variations of reported GHG emissions for excavation and disposal for example could be due to some form of soil treatment before final disposal included in a particular data set for one study, while another study may not have had it included. It is important therefore that authors investigate the data used in their studies and report them accurately.

More of the evidence however, from these different life cycle inventories resulting in differing GHG estimates is represented in Lemming et al., 2010a,b,c, which reviews the different effects of using different LCI data. Volkwein et al., 1999 also compared a number of remedial options for specific contaminated sites. In that study, three very different conclusions ranging from 0.04 t CO₂-eq/m³, site treated to 0.25 t CO₂-eq/m³ offsite treated, all stemming from the particular life cycle stages involved in the specific remediation process.

5.3. Comparison with in-situ soil treatments

According to Kuppusamy et al., 2016), the environmental impacts of ex-situ treatment occur mainly on a global scale, while the impacts of in-situ remediation are both local (the concentrations remaining in the soil are generally more uncertain for in-situ treatment) and global (energy use). For offsite treatments, since the transport of soil stage in the life cycle of the treatment is the principal cause of environmental impact, the distance of the transport is a key factor in the choice between in-situ and ex-situ offsite treatment (Federal Remediation Technologies Roundtable (FRTR), 2002). According to Ribbenhed et al., 2002) and Vignes, 1999, when the environmental impacts of ex-situ treatment techniques are compared, thermal treatment has the highest global warming potential; and usually has the highest remediation efficiency as well. Soil washing often records low GWP and little

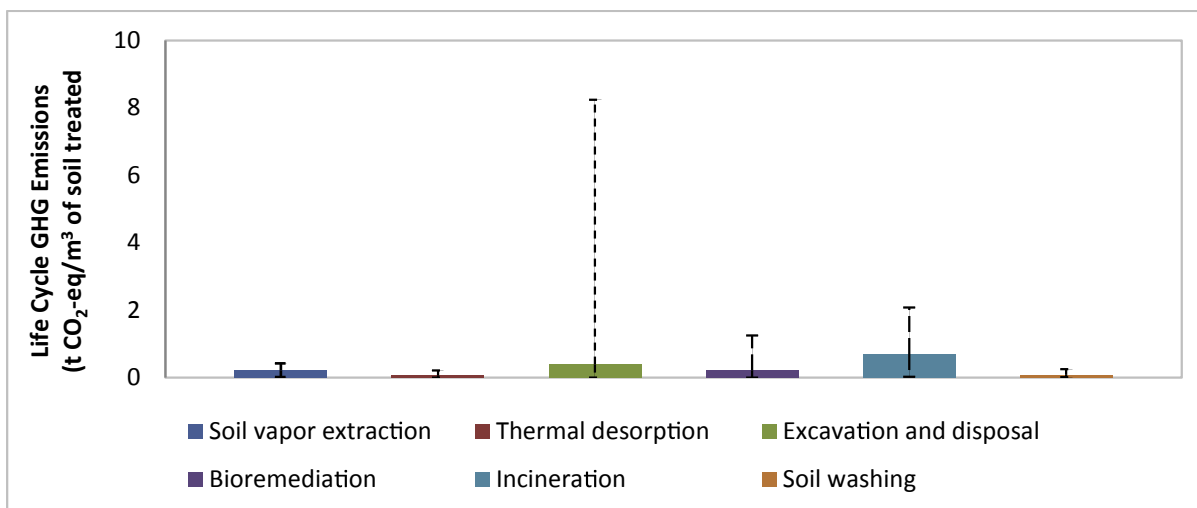


Fig. 2. Life cycle GHG emission estimates of ESRTs.

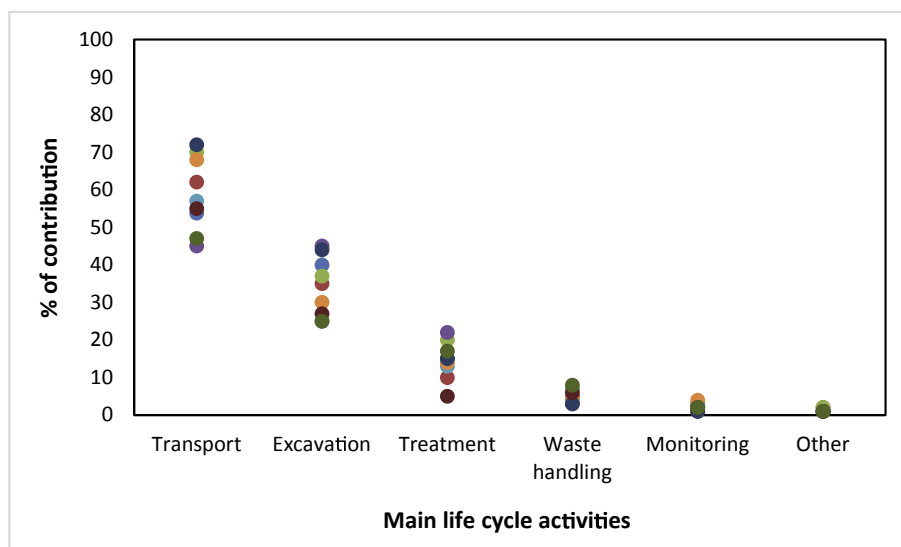


Fig. 3. Life cycle greenhouse gas emissions contributions by life cycle stage.

cleaning effect (Diamond et al., 1999), (Ribbenhed et al., 2002).

In-situ technologies however, often involve applying chemical, biological, or physical processes to the subsurface to degrade, remove, or immobilize contaminants without removing the bulk soil (Camenzuli et al., 2013). Compared to excavation and ex-situ treatment, the use of these technologies offers several benefits, such as addressing deep contamination and generally costing less. The main advantage of *ex-situ* treatment is that it generally requires shorter time periods than *in-situ* treatment, and there is more certainty about the uniformity of treatment because of the ability to homogenize, screen, and continuously mix the soil. However, since *ex-situ* treatment requires excavation of soils, it often leads to increased costs and engineering for equipment, possible permitting, and material handling/worker exposure considerations as well as higher energy requirements often leading to higher GHG emissions. In contrast, *in-situ* remediation techniques require less energy, create less waste and in most cases generate fewer greenhouse gas emissions (F. of C. M. FCM, 2008). Fig. 4 shows comparison between *ex-situ* soil remediation technologies (ESRTs) and *in-situ* soil remediation technologies (ISRTs). Data used in Fig. 4 represents the worst-case scenarios (maximum emission values) for ESRTs from sources of this review and average values for selected ISRTs. The life cycle emissions are comparatively higher in ESRTs ($8.2 \text{ t CO}_{2\text{eq}}/\text{m}^3$) as compared to ISRTs ($3 \times 10^{-4} \text{ t CO}_{2\text{eq}}/\text{m}^3$) from the studies reviewed. The wide variation in GHG emissions of ESRTs could be due to different management options for the different contaminant types. Some studies including (Songet al, 2017; Tajam et al., 2010; Wicket al, 2016) have suggested that some *in-situ* thermal treatments could contribute significantly higher GHG emissions depending on the electricity/heat source mix.

5.4. Life cycle GHG reduction strategies for *ex-situ* remedial options

Since the technique of life cycle assessment accounts for impacts (environmental) at all stages in the treatment process, a full life cycle assessment of an ESRT (including extraction of materials to receiving a remediation certificate in Alberta, Canada for example) can identify specific stages in the treatment life cycle that contributes the most to GHG emissions. This information can be very important in developing strategies and plans to reduce GHG

emissions associated to the different phases of the remediation technology. The focus on *ex situ* technologies is important, as ESRTs, especially excavation and disposal is often required at contaminated sites to address immediate risks. Moreover, there may be instances where *in situ* technologies may be infeasible due to technical constraints, long duration or associated costs. According to the United States EPA green remediation program (USEPA, 2008a), many opportunities exist to reduce the negative impacts of ESRTs. Primarily, Best Management Practices (BMPs) can be adopted to help reduce energy (usually fuel) consumption, which is the primary source of GHG emissions during soil treatment. Some of these strategies may include choices to reduce transportation, such as selection of offsite treatment or disposal site, nearest laboratory for analyses, alternative means of transport, amongst others, as specifically outlined in the USEPA program.

5.5. Additional life cycle impacts of ESRTs

In this article, we specifically review Life cycle greenhouse gas estimates. However, an LCA should, fundamentally, include all relevant potential environmental impacts related to the goal of the study (Turconi et al., 2013). There are many other environmental impacts of soil remediation technologies beyond the emission of greenhouse gases. In addition to global warming, acidification and eutrophication are other important impact considerations associated with soil remediation technologies (Lemming et al., 2010a). Another important environmental burden, which is often not considered by most remediation LCAs, is noise (Turconi et al., 2013). This could often be associated with the phase of soil excavation, which involves large equipment and machinery that are often accompanied with large noise during their operation. Again, the risks associated with long-term storage of contaminated soils at landfills to the local ecosystems and biodiversity for such options may be considered. There could also be the long-term potential for induced risk of seismic effects due to excavation of contaminated soil and extraction of fossil fuels usually associated with the upstream processes.

The choice of impact categories selected to be evaluated based on the goal of the LCA study can heavily affect the conclusions of the study. While one only focuses on GWP in a study where a remediation option performs well in that regard, it may perform

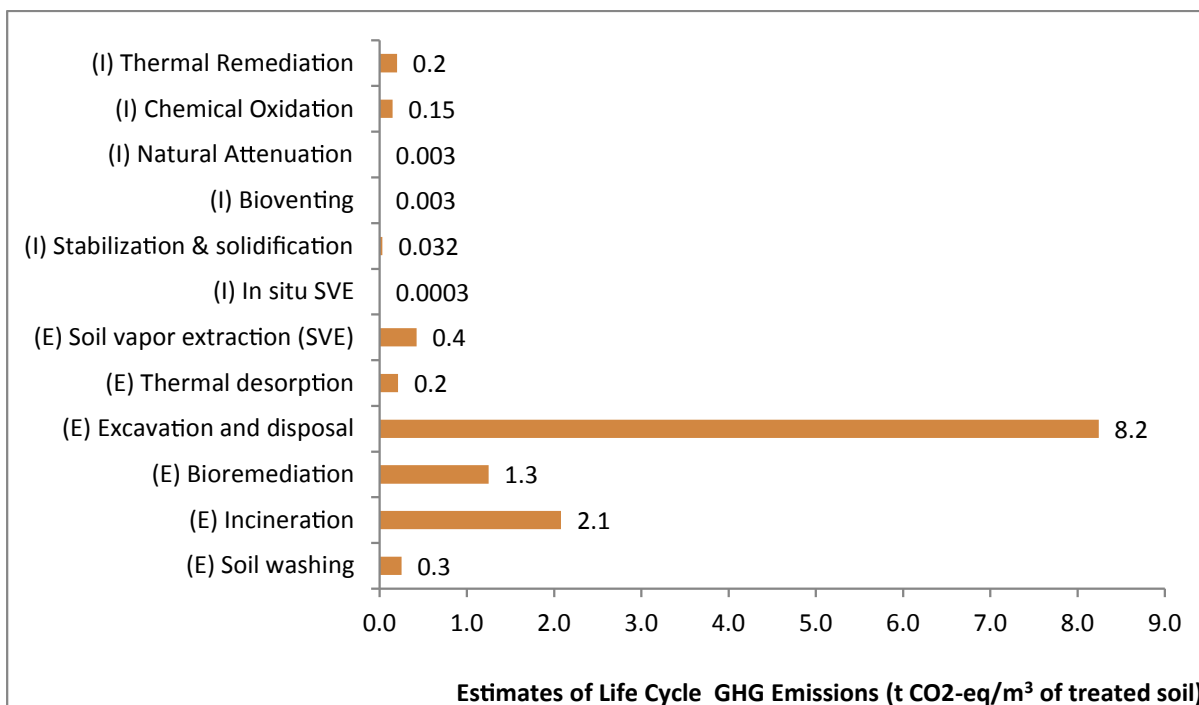


Fig. 4. Maximum life cycle GHG emission levels of soil remediation technologies.

poorly in ecotoxicity for example. This may lead to a certain conclusion. However, since LCA can be used to compare remediation scenarios in terms of their associated environmental burden, it could be useful to consider several other impact categories, to fully make conclusions on the suitability of a particular remediation technology in terms of their sustainability. As such, a full LCA, which considers several other impact categories, may be more effective in selecting a sustainable technology for remediation.

5.6. Opportunities for improvement in LCA remediation studies

This current study like many other LCA studies often show the effectiveness of life cycle analysis in assessing the environmental impacts of remediation technologies – ex-situ soil remediation technologies in this case. However, due to the fact that LCA is a data-intensive exercise, the quality of data is paramount and has a significant impact on the accuracy, level of uncertainty and credibility of the modelling output (Amponsah et al., 2014). As such, any form of data inaccuracies must be appropriately dealt with. One opportunity for improving such challenges is to include clear statements of data sources, considered assumptions and a clear definition of the system boundaries under investigation. This provides an opportunity for further research in developing a framework to guide LCA practitioners in applying the LCA tool effectively to site remediation to help reduce uncertainty, increase harmonization and improve transparency of results.

6. Conclusion

This study reviewed and collated literature relating to life cycle greenhouse gas emissions associated with different ESRTs. We compiled and analyzed GHG emissions data from published LCA studies and the variability of existing LCA GHG emissions in six ESRTs. While some of these differences may reflect actual differences in GHG emissions, others may largely be due to assumptions, system boundary, LCA modelling choices, and different LCA

approaches, including selected functional units, methods and software, geographic location and processes. The results showed that the highest mean GHG emission value, 0.7 t CO₂-eq/m³ was associated with incineration, whilst the lowest, 0.07 t CO₂-eq/m³ was associated with thermal desorption. It was also found that there was a large variation range of GHG emissions from the ex-situ excavation and disposal treatment technologies, varying from 3.1×10^{-7} t to 8.2 t CO₂-eq/m³ of treated soil. We compared the GWP levels of ex-situ remediation methods to the GWP levels of typical in-situ remediation. The life cycle emissions of ESRTs were higher (8.2 t CO₂eq/m³) than that of ISRTs (3×10^{-4} t CO₂eq/m³). The study reaffirms the need for LCA studies to be more transparent in disclosing the assumptions, method and boundary of the study. This will definitely improve the quality of LCA emissions inventory and user confidence in LCA calculations related to contaminated site treatment technologies.

The results of this review can provide suitable baseline estimates for future projects in developing sustainable contaminated soil remediation technologies in order to mitigate GHG emissions. As cleanup technologies continue to advance and incentives evolve, such data can be essential in offering significant potential for increasing the net benefit of cleanup, saving project costs, and expanding the long-term use or re-use of property. The use of such data can support green remediation practices, reducing the demand placed on the environment during cleanup actions, and avoid the potential for collateral environmental damage. Furthermore, this knowledge provides opportunities to increase sustainability throughout the investigation, design, construction, operation, and monitoring phases of site remediation regardless of the selected cleanup remedy. Therefore, the LCA GHGs are a useful tool to assess the impact of different scenarios and management practices on the ESRTs.

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